EMISSIVITY MEASUREMENTS ON METALLIC SURFACES WITH

VARIOUS DEGREES OF ROUGHNESS: A COMPARISON OF LASER

POLARIMETRY AND INTEGRATING SPHERE REFLECTOMETRY[1]

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ABSTRACT

Both integrating sphere reflectometry as well as laser polarimetry have their advantages and limitations in their ability to determine the normal spectral emissivity of metallic samples. Laser polarimetry has been used for years to obtain normal spectral emissivity measurements on pulse-heated materials [1, 2]. The method is based on the Fresnel equations, which describe reflection and refraction at an ideally smooth interface between two isotropic media. However, polarimetry is frequently used with surfaces that clearly deviate from this ideal condition. Questions arise with respect to the applicability of the simple Fresnel equations to non-specular surfaces. On the other hand, reflectometry utilizing integrating spheres provides a measurement of the hemispherical spectral reflectance, from which the normal spectral emissivity can be derived. Integrating-sphere reflectometry provides data on spectral-normal-hemispherical reflectance and hence normal spectral emissivity for all kinds of surfaces. However resulting errors are minimal when both the sample and the reference have a similar bidirectional reflectance distribution function (BRDF). In an effort to explore the limits of polarimetry in terms of surface roughness, room temperature measurements on the same samples with various degrees of roughness were performed using both an integrating-sphere reflectometer and a laser polarimeter. In this paper the two methods are briefly described and the results of the comparison are discussed.

Keywords: emissivity; integrating sphere; laser polarimetry; reflectometry; rough surfaces; roughness.

1. INTRODUCTION

In a variety of high speed, high temperature experiments pyrometry is the only way to obtain reasonable temperature data. Such experiments are pulse heating calorimetry to get thermophysical properties of metals and alloys in the solid [3, 4] and liquid [1, 2] state, shock physics experiments to get equations of state of metallic samples [5] and levitation experiments to get surface tension and data of supercooled liquids [6], just to name a few. At the Los Alamos National Laboratory shock compression experiments are routinely performed to obtain the equations of state of materials. The typical duration of such experiments is about 1 to 5 μs, which is why pyrometry is well suited to obtain temperature data.

Using a pyrometer that is calibrated in terms of blackbody radiance, the radiance or blackbody temperature (at the mean effective wavelength of the pyrometer) can be measured very accurately [7], but to obtain thermodynamic- or true temperatures from this radiance temperature, an additional quantity, the normal spectral emissivity (also at the mean effective wavelength of the pyrometer), has to be known. There are several different ways to determine the normal spectral emissivity, i.e. laser polarimetry [1], integrating sphere reflectometry [3], comparing the thermal radiation emitted from the surface of interest to the thermal radiation emitted from a blackbody cavity at the same temperature [4], or measuring the absorptance ratio at two wavelengths [8], just to mention the more common.

In the shock-physics community, multi-wavelength pyrometry has widely been used to determine the temperature and normal spectral emissivity of shocked samples [9]. Recently, efforts have been underway to utilize reflectometry (either with or without

integrating spheres) [10, 11] and laser polarimetry [5] in these experiments, to

dynamically measure emissivity and improve the accuracy of the temperature

measurements.

The contactless character of laser polarimetry is a very convenient feature in the face of

the highly destructive nature of shock-compression experiments, where any equipment

that is placed close to the sample is usually severely damaged. Its weakness lies in the

constraints that it places on the sample surface in terms of texture and the presence of

oxide layers. The insensitivity of integrating sphere reflectometry to these factors is

this technique's greatest advantage.

1.1. Laser polarimetry

Laser polarimetry is based on the measurement of the change in the state of polarization

of a laser beam upon reflection at the surface of a sample. Figure 1 shows a schematic

of the measurement geometry.
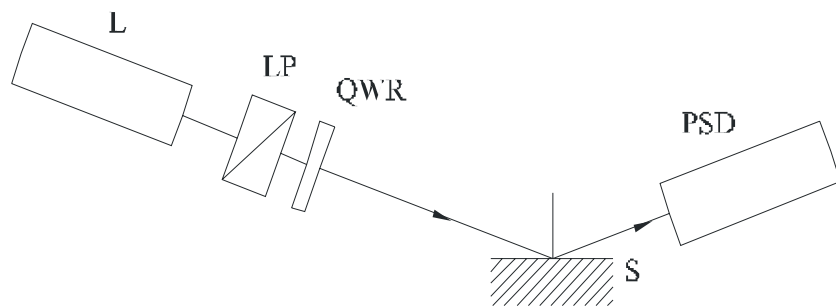
Fig. 1. Schematic drawing of a laser polarimeter. L, laser; LP, linear polarizer; QWR,

quarter-wave retarder; S, sample; PSD, polarization state detector.

From the measured change in polarization the index of refraction n and extinction

coefficient k of the sample can be derived using the Fresnel equations [12]. These

equations describe the reflection of an optical plane wave at the planar interface

between two isotropic media in terms of the amplitudes and phases of the incident and

reflected waves.

Making use of Kirchhoff's law in the energy balance between the incident, absorbed,

and reflected light fluxes at the surface of an opaque sample, the normal spectral

emissivity of the sample is finally obtained from:

$$\varepsilon = \frac{4n}{(n+1)^2 + k^2} \tag{1}$$

with ε the normal spectral emissivity. This equation is only valid for vacuum (or air)-

to-metal interfaces. More details about data reduction and the application of laser

polarimetry to measurements on pulse-heated samples can be found in Refs. [1] and

[2].

1.1.1. Problems with laser polarimetry

The validity of the Fresnel equations, and hence the applicability of laser polarimetry,

is limited to ideally smooth surfaces for which reflection is perfectly specular. The

difference between reflection from a planar and a rough surface is illustrated in Fig. 2.
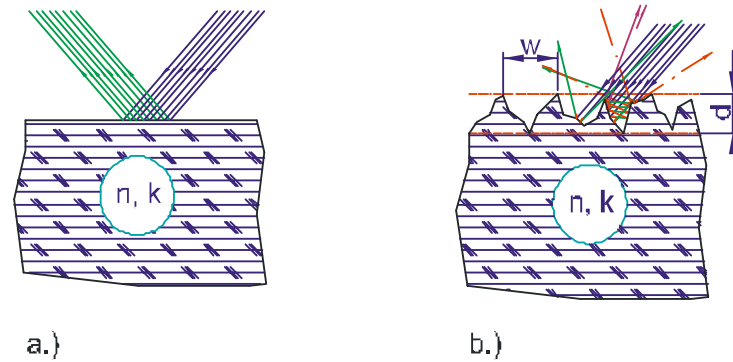
Fig. 2. Reflection of a light beam at (a.) a smooth and (b.) a rough surface. The blue rays indicate incident light, green indicates single reflection, and red multiple reflections.

At a rough surface multiple reflections can occur, which, among other things, will lead to a depolarization of the reflected beam and render the use of the Fresnel equations impossible. Up to now there have been no satisfactory procedures developed to apply laser polarimetry to the measurement of optical properties of rough surfaces. If laser polarimetry were feasible to determine optical parameters of rough surfaces, additional information about surface topology would be required to get the emissivity. As surface conditions change very rapidly over a wide range at the above-mentioned experiments, an exact formalism (taking everything into account) to determine emissivity using laser polarimetry on dynamic experiments under such demanding conditions seems not to be in sight. The scope of this paper is to investigate the effect of surface roughness on the results of laser polarimetry.

The other drawback of laser polarimetry is that the polarimeter has to be aligned very accurately and any sample movement, tilt or bending misaligns the setup and large errors can occur [13].

1.2. Integrating sphere reflectometry

As the name implies, this technique employs an integrating sphere to collect laser light that is reflected by the sample into the entire hemisphere above it. Multiple reflections on the highly and diffusely reflecting inner surface of the sphere distribute the light uniformly over the entire sphere in a short time. It is a relative measurement, where the signal obtained using the sample is referenced to that obtained using a reflectance standard, whose reflectance is accurately known. In the ideal case, the ratio of the radiances produced inside the sphere is equal to the ratio of the reflectances of the sample and the standard. Consequently,

$$\frac{S_s}{S_r} = \frac{R_s}{R_r}.$$

(2)

Here S_s is the signal obtained with the sample, S_r is the signal obtained with the reference, R_s is the reflectance of the sample, and R_r is the reflectance of the reference material.

For opaque samples, the normal spectral emissivity is obtained from the measured spectral hemispherical reflectance for normal incidence using Kirchhoff's law in the energy balance between the incident, absorbed, and reflected light fluxes at the sample surface:

$$\varepsilon = 1 - R_s$$

(3)

Here ε is the normal spectral emissivity of the sample and R_s is the spectral normal-hemispherical reflectance of the sample.
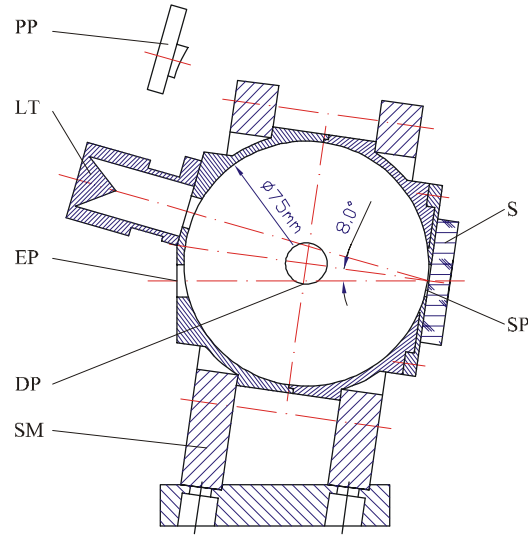
Fig. 3. Schematic of the integrating sphere reflectometer. PP, port plug; LT, light trap; EP, entrance port; DP, detector port; SM, sphere mount; S, sample; SP, sample port.

A schematic drawing of a simple integrating sphere reflectometer is shown in Fig. 3. It can be seen that in order to prevent the specularly reflected component from exiting the sphere after only one reflection, the entrance port is not located directly above the sample, but at a small angle off the normal. This is usually neglected when Eq. 3 is applied.

More details on integrating sphere reflectometry as applied to dynamic emissivity measurements on pulse-heated samples can be found in Ref. [3]. Ref. [14] gives a detailed treatment of integrating sphere theory.

1.2.1. Problems with integrating sphere reflectometry.

Although integrating sphere reflectometry is not very sensitive to surface roughness and contamination, many things have to be considered in order to get reliable results. In Ref. [15] the following main errors are listed: a) errors due to lack of perfect

diffusion of light reflected from the sphere walls; b) those due to unequal illumination of test and standard surfaces when the substitution method is used; c) loss of light through the necessary holes in the sphere wall; d) when the test surface is viewed directly by the photometer, the brightness in the viewing direction may depend upon the diffusion characteristics of the test surface and may not be directly proportional to the reflection factor of the surface; e) obstruction of light by an opaque screen, if used, and f) errors due to stray light not confined within the concentrated beam projected into the sphere. According to our experience the error due to different reflection characteristics of the sample and the reference used (d) in combination with imperfect lambertian reflectance at the sphere walls (b) are the main contributors (also in spheres carefully engineered for a special application) to measurement uncertainties in our case. Rough estimates suggest this error may be up to 8 percent, which is in good agreement with a calculation in Ref. [16]. In this reference the sources of the error are discussed in detail. In correcting this error the BRDF need not be known, only the percentage of light which is reflected specularly [17]. Knowing the samples specularity (%spec.) and using both a diffuse and a specular standard, the reflectance is obtained by:

$$R_s = S_s \left(\%spec \cdot \frac{R_{s,r}}{S_{s,r}} + \left(100 - \%spec\right) \cdot \frac{R_{d,r}}{S_{d,r}} \right). \tag{4}$$

Here $R_{s,r}$ is the reflectance of the specular standard, $S_{s,r}$ is the signal measured with the specular standard, $R_{d,r}$ is the reflectance of the diffuse standard, and $S_{d,r}$ is the signal measured with the diffuse standard. This equation takes the deviation of the sample reflection from ideal lambertian behavior into account and corrects most of the error.

2. MEASUREMENTS

2.1. Instrumentation

A Division-of-Amplitude-Photopolarimeter (DOAP) was used for the polarimetric measurements. It is a commercial instrument that was purchased from Containerless Research (CRI) and has since been upgraded to more closely meet our demanding field requirements. It is a high-speed instrument capable of measuring emissivity every 20 ns. A laser diode that is used in series with an Erbium-Doped-Fiber-Amplifier (EDFA) serves as the light source and can deliver up to 2 W at 1.55 µm. The output power level can be adjusted continuously by varying the current through the laser diode. A detailed discussion of this particular instrument can be found in Ref. [5]. More detail about DOAPs in general can be found in Refs. [18-20].

For the spectral-hemispherical reflectance measurements we built a simple reflectometer centered on a 70 mm diameter, gold-coated integrating sphere. The sphere, which was purchased from Gigahertz Optik in Germany, features four circular ports whose total area is less than 5% of the sphere surface area to ensure high sphere efficiency. The same laser that was used with the polarimeter was also used with the reflectometer. A collimated and mechanically chopped laser beam passed through the entrance port and illuminated the sample or the reflectance standard at an angle of 8° off of normal. After spatial integrated by the sphere, the reflected light was detected by an InGaAs detector fiber-coupled to the detector port of the sphere. Either a light trap or a port plug (having the same properties as the sphere inner surface) can be attached on the fourth port which is where the laser beam is reflected on a specular sample, in order to estimate the specularity of the sample (see Fig. 3).

2.2. Samples

Measurements were performed on eight samples each of copper, tin, aluminum 1100 (>99.0 weight% Al, 0.12 weight% Cu) and aluminum 6061 (97.9 weight% Al, 0.60 weight% Si, 0.28 weight% Cu, 1.0 weight% Mg, 0.2 weight% Cr). The samples were 40 mm diameter discs with a thickness of 6.4 mm. For each material one sample was polished, one was diamond turned, and six had surfaces with various degrees of roughness. Of the six rough samples, half were "randomized" and the other half "non-randomized". These two types correspond to what is often referred to in literature as samples with "randomly rough" and "contoured" surfaces, respectively. The randomized samples had their surfaces treated with abrasive paper, whereas the non-randomized were left with surface tooling marks. Since they were fabricated on a lathe, these marks were circular, as illustrated in Fig. 4, and were expected to give rise to a grating effect which, if true, would be detrimental to polarimetry.
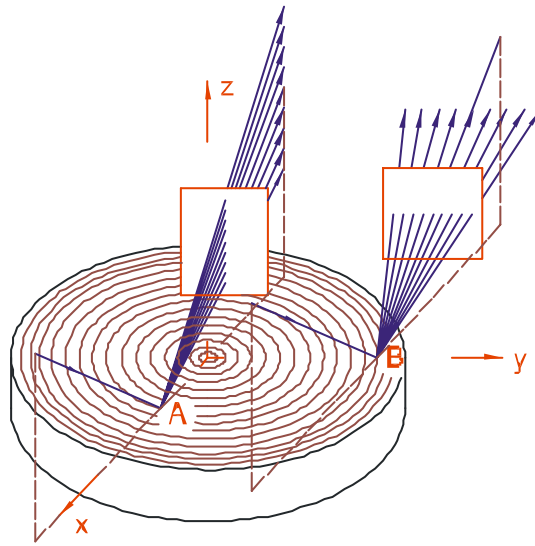
Fig. 4. Reflection on a structured sample.

The three samples in each group (randomized and non-randomized) had surfaces with

an RMS-roughness of 8 μinches, 16 μinches, and 32 μinches. These values were

measured using a stylus profilometer (Federal Products Surfanalyzer 5000®).

With each sample, measurements were taken at three locations with the polarimeter and

at five locations with the reflectometer. The measurements taken with each method

were averaged and a standard deviation was computed. Each polarimeter measurement

itself represents an average of 4096 data points.

2.3. Estimation of the specular component.

As reference samples we used a set of 8 diffuse reflectors with a reflectivity between

99 and 2% and a lambertian reflectance distribution, purchased from Labsphere, Inc. as

diffuses standards, and a polished copper disk, which served as a specular standard.

The reflectivity of this copper disc was assumed to be 96.6% (according to Ref. 21).

The reflectance values of the diffuse standards could be reproduced with an uncertainty

of less than 1% using the 99% standard as reference. However using the diffuse

reference and measuring the reflectance of the specular copper disc led to an

uncertainty of about 8%, as mentioned earlier. This in turn led to the error correction

discussed above. Reference 22 describes a standard method for describing the

specularity of a reflectance sample using an integrating sphere. This method requires a

special sphere designed specifically for the beam geometry used at the experiment.

Instead we assumed the diffuse Labsphere standards to be ideal lambertian reflectors

and compared the fraction of light reflected by the sample escaping through the

specular reflectance port (with the attached light trap) to the fraction of light escaping

through the same port when light was reflected by the diffuse Labsphere standard. The

specularity of the sample (%spec) was then calculated using:

$$\%spec = 1 - \frac{S_{s,LT}/S_s}{S_{d.r.,LT}/S_{d.r.}} \ . \tag{5}$$

Here $S_{s,LT}$ is the signal measured with the sample and the specular light trap and $S_{d.r.,LT}$

is the signal measured with the diffuse reference and the specular light trap. Using eq.

5 a specularity of 0% was assigned to the diffuse standards and of 100% to the copper

disc. The values for the different samples are in between (see Tab. I).


3. RESULTS

The values for the measured specularities for all samples investigated in this study are

given in Table I. Table II shows the value for the measured reflectivity using both

methods, also for all samples investigated.

Table I.   The measured specularities, according to Eg. 5, of the different samples

used in this investigation

surface type	Cu	Sn	Al1100	Al6061
polished	100		100	100
diamond turned	100	97.1	100	100
$8\,\mu$inch / randomized	9.5	59.5	18.3	20.5
$16\,\mu$inch / randomized	4.2	4.7	6.6	9.8
$32\,\mu$inch / randomized	4.1	7.7	4.0	4.9
$8\,\mu$inch / not randomized	16.4	13.0	21.5	54.1
$16\,\mu$inch / not randomized	15.9	15.2	17.4	31.7
$32\,\mu$inch / not randomized	5.2	18.3	6.9	8.3

Table II. Reflectance (in %) at 1.55µm for four different materials with various degrees of surface roughness, as measured with the integrating sphere reflectometer (ISR) and the division-of-amplitude-photopolarimeter (DOAP)

surface type	Cu		Sn		Al 1100		Al 6061	
	ISR	DOAP	ISR	DOAP	ISR	DOAP	ISR	DOAP
polished	97.1	94.42		91.4	93.8	93.4	87.4	
diamond turned	96.6	95.76	82.6	85.7	79.4	73.0	91.8	91.1
8 µinch / randomized	69.4	50.4	61.1	66.3	88.2	68.7	78.7	67.4
16 µinch / randomized	87.1	82.8	67.7	58.8	76.6	67.0	86.6	54.0
32 µinch / randomized	82.9	63.7	78.7	74.0	88.5	65.4	84.5	69.6
8 µinch / non-randomized	97.2	80.6	76.5	71.8	93.3	73.2	92.2	87.9
16 µinch / non-randomized	89.2	80.4	76.2	52.0	89.4	76.3	91.3	87.5
32 µinch / non-randomized	94.1	95.1	79.7	82.1	91.7	93.8	88.9	86.7
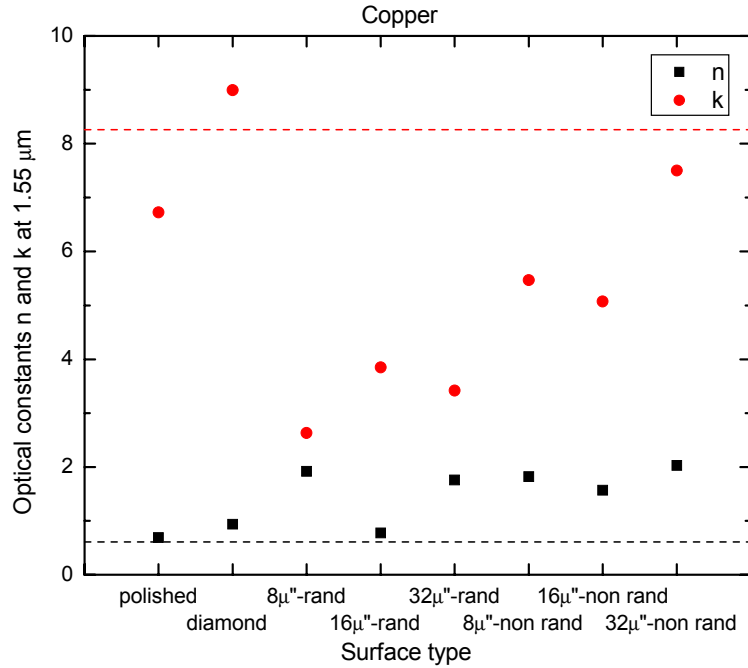
Fig. 5. The index of refraction n and extinction coefficient k as measured with our polarimeter on the copper samples. The dashed lines represent values from Ref. [21]

The fact that the optical parameters n and k can be measured accurately by polarimetry only for optical smooth surfaces can be seen from Fig. 5, which shows this parameters as a function of the surface condition for copper. It is evident that the deviation from published values (dashed lines) is very large, also for small surface roughness. Nevertheless, as can be seen in Fig. 6, the calculated emissivity value (using eq. 1) is not very sensitive even to large errors in the optical parameters. The values assumed to calculate the emissivity in Fig. 6 are in the range of the measured values according to Fig. 5.
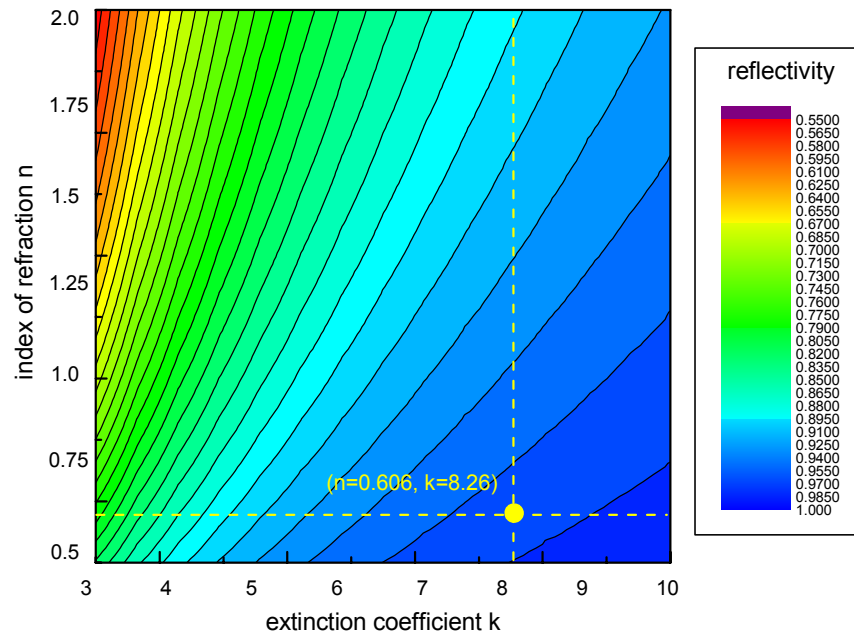

Fig. 6. Reflectivity calculated using Eg. 1, with varying optical parameters n and k. The yellow spot indicates the literature value [21] for copper at room temperature

One can see that for all experimental values of n and k the calculated reflectance is too low and hence the emissivity is too high. On the other hand, multiple reflections occur on rough surfaces (see Fig. 2), which means that the effective reflectivity ρ_{eff} decreases to:

$$\rho_{eff} = \rho_0^{\bar{n}} \tag{6}$$

where ρ_0 is the reflectivity of the smooth surface and $\bar{n}$ is the average number of reflections. Since both effects oppose each other, the results on randomized surfaces obtained by polarimetry are therefore in good agreement with those obtained by reflectometry. These results are plotted for tin in Fig. 7.
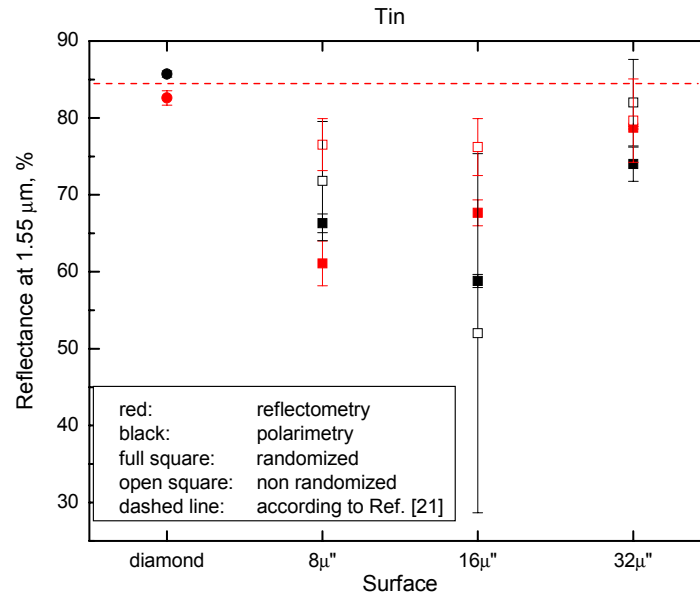
Fig. 7. Measured reflectance of tin on samples with various degrees of roughness. The error bars indicate the standard deviation of the measurements.

For not randomized surfaces the agreement between both types of measurements are much worse, which can be explained by the reflectance characteristic of a surface with tooling marks, as shown in Fig. 4. Depending on the position where the beam is reflected from the sample, a grating effect occurs in different directions. This effect has great impact on both measurement methods, which can be seen on the large standard deviations on these surfaces.

The uncertainty in the polarimetric emissivity measurements on polished and diamond turned samples is believed not to exceed 3% [1]. In the case of measurements on rough samples an uncertainty may not apply, since the technique itself might not be applicable. The largest deviation of the polarimetry results from the measurements obtained using the reflectometer was 4% for the randomized samples and 20% for the non-randomized samples.

We have not yet conducted a rigorous uncertainty analysis of the reflectometry measurements. Using the standard deviation of the measurements at different sample locations as a rough estimate, we believe that the uncertainty does not exceed 5% and 8% for the randomized and non-randomized samples, respectively.

4. CONCLUSIONS

As expected, the measurements obtained with polarimetry and reflectometry on the polished and diamond turned samples generally agreed well with each other and with the literature. However, in the case of the aluminum samples the agreement was not very good. It is not clear why this is true. It is conceivable that the disagreement was caused by the fact that the samples were not of pure aluminum and that the effect of surface oxides was larger in the case of the aluminum samples.

Reflectometry and polarimetry were also in reasonable agreement in the case of the randomized samples, as can be seen in Tab. II and Fig. 7 for tin. Interestingly enough, the polarimetry results followed those obtained with the reflectometer as the surface type changed, even though in this case it is not clear what the physical meaning of the measured n and k values is.

This was not true in the case of the non-randomized samples. The polarimetric measurements deviated significantly from those obtained with the reflectometer. We suspect that this was related to the grating effect that is caused by the periodicity in the roughness of these samples (see Fig. 4). Probably for the same reason, the standard deviation between polarimetric measurements at different locations on the non-randomized samples was significantly larger than in the case of the randomized samples. This is expected because the polarization of the reflected beam strongly depends on the orientation of the periodic grooves on the sample with respect to the plane of incidence.

More research will be required to really understand our observations. It can already be stated that polarimetric emissivity measurements on anything other than specular surfaces should be treated with caution. It seems to be the case that randomly rough surfaces are more forgiving than contoured surfaces in this respect. Even though reflectometry utilizing integrating spheres is better suited for measurements on rough surfaces, there are still a considerable number of potential sources of error relating to the characteristics of the sphere and their deviation from ideal behavior. A combination of diffuse and specular reflectance standards can certainly reduce some of these errors to the point where reflectometry becomes a viable option for use in shock experiments.

ACKNOWLEDGEMENTS

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